Journal of Power Sources 247 (2014) 384–390

Contents lists available at ScienceDirect

Journal of Power Sources

journal homepage: www.elsevier.com/locate/jpowsour

Repetitive hot-press approach for performance enhancement of hydrogen fuel cells

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HIGHLIGHTS

- Confirms the efficacy of the hot-pressing technique under optimal conditions.
- Two stage hot-press with hydration between presses was found to improve performance.
- Gains from hot-pressings due to the kinetic polarization suppression.
- A correlation between water content and fuel cell performance was investigated.

ARTICLE INFO

Article history:
Received 8 May 2013
Received in revised form
24 August 2013
Accepted 26 August 2013
Available online 3 September 2013

Keywords:
PEMFC
Nafion
Hot-press
Membrane electrode assembly

ABSTRACT

For Nafion 212-based PEM fuel cells assembled with gas diffusion electrodes, two stage hot-press with conditioning or hydration between presses was found to improve performance and to allow the cell to sustain higher current densities. Testing refuted additional time as a driving factor for increased performance and the optimal hot-press time of 3 min from literature is confirmed. The two-stage hot-press process is shown to decrease ohmic resistance and to increase performance by a greater factor for Nafion 212 than Nafion 115. Tafel plots are constructed to focus on kinetics and give insight into the behavior of each membrane. Gains from hot-press and sequential hot-pressings are shown to be largely due to the suppression of kinetic overpotential. A correlation between water content and fuel cell performance in hot-press processes indicates that excessive hot-press limits the water content of a membrane, lending further support to an existence of an optimal hot-press time.

1. Introduction

Proton exchange membrane fuel cells (PEMFC) that use hydrogen gas as fuel have received considerable attention over the past two decades due to their high energy density and low emissions [1]. Fuel cells can power vehicles, provide stationary power, reduce dependence on foreign oil and reduce air pollution. Optimization of these cells can make them a more viable alternative energy source by improving their energy density and cost [1,2]. At the heart of the PEMFC is the membrane electrode assembly (MEA) which is composed of electrocatalyst layers separated by an ion-conducting membrane. MEA hot-pressing is an important issue in fuel cell performance, as the process can help solidify the structure of the MEA and increase electrical contact [3–7]. Additionally, hot-pressing can lead to favorable polymer reconfiguration and catalyst utilization to benefit fuel cell performance [8,9]. However, excessive hot-press can lower performance by irreversibly drying out and damaging the MEA, starting at temperatures above 130 °C [2]. Tashima et al. show this damage with X-ray Photoelectron Spectroscopy and note that the CF_2/CH and CF/CH ratios decrease at 150 °C, evidencing main chain structure decomposition [2]. SEM imaging at excessive hot-press conditions showed the catalyst layer had been forced into the conductive layer and covered by Nafion, reducing the surface area and is a likely reason for decreased performance [3]. Sone et al. propose that the thermal treatment of Nafion 117 can reduce its water content through the reduction of micropores which proportionally reduces the conductivity of the cell [10]. Membrane water content is an important indicator of fuel cell health and performance, since protons are transported through the membrane while attached to water as hydronium ions [11]. A water-swollen pore increases the proton diffusion through the membrane due to the Grotthuss mechanism [12]. Studies for hot-press optimization have found pressures between 6.90 and 7.57 MPa, temperatures between 99 and 132 °C, and durations of

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http://dx.doi.org/10.1016/j.jpowsour.2013.08.090

2–3 min as most beneficial [2,3,13]. Other reported hot-press processes are performed between 3 and 7.57 MPa, temperatures between 99 and 135 °C, and for 1.5–3 min [4,6,14–17].

This work will analyze the effect of hot-press and repeated hot-press on fuel cell performance, the effect of conditioning, and water hydration. Hot-press techniques will be investigated, including sequential hot-presses, and their effect on the MEA will be noted.

2. Experimental

2.1. Fuel cell tests

A Scribner 850e fuel cell test system was used in our experiments. Temperatures were set to 60 °C for the cell and the anode and cathode humidifiers to ensure 100% relative humidity. The flow rates of hydrogen and oxygen were 100 sccm, while back pressure was regulated at 15 psi. Each MEA was conditioned before its first test at a constant 0.6 V and 50 sccm flow rate until the maximum attainable current developed. This typically took 12–24 h. In assembly, the test fixture was tightened to 6 Nm of torque on each of the eight bolts in the cell frame. A model 4386 Carver hot-press equipped with a digital pressure gauge was used for hot-pressing the membranes at a temperature of 132 °C and a force of 300 lb_f (1335 N). Membrane thickness was measured before and after presses with a caliper with an accuracy of ±0.0005 in (0.0127 mm).

Hand sprayed, conventional gas diffusion electrodes (GDEs) were fabricated. AVcarb™ GDL was uniformly sprayed with a 50% Pt/C ink dispersed in a water/IPA/liquid ionomer mixture in 5 min increments until the target loading of 0.1 mg cm^{-2} of platinum was reached on the GDL. The GDL was cut into 1 cm^2 squares for the construction of the MEA.

2.2. Membrane water content tests

1 cm × 3.5 cm rectangular strips were cut from a sheet of Nafion 212. After the appropriate hot-press procedure, the membranes were placed in separate, labeled, open, glass containers with silica gel beads to aid the dehumidification of the Nafion. These were then dried in a Yamato oven at 80 °C for three days. Five total measurements of the dry weight of the Nafion strip were taken after an initial measurement; one after each passing hour. The membranes were placed back into the oven between measurements. After the conclusion of the dry weight measurements, the strips were placed in water and allowed to soak on a hot plate at 80 °C for 2 h. After soaking, the membranes were left in water for five days at room temperature to fully hydrate. For the wet weight measurements, the Nafion strips were removed from the water containers, wiped until no visible moisture could be seen, weighed, and then returned to the water containers. Ten measurements of each membrane were taken and outlier values were discarded. The values are compared using the equation for water content [11],

$$W_c = \frac{100\%\ (W_w - W_D)}{W_D} \tag{1}$$

where W_w and W_D are the respective wet and dry sample weights. The standard deviation was calculated by taking into account the standard deviations of the dry weight and wet weight, as described by D.C. Baird [18]:

$$\sigma = \sqrt{\left(\frac{\partial W_c}{\partial W_w}\right)^2 \sigma_{W_w}^2 + \left(\frac{\partial W_c}{\partial W_D}\right)^2 \sigma_{W_D}^2} \tag{2}$$

3. Results and discussion

3.1. Effects of hot-press on the power and polarization behavior for N212 and N115 based cells

Hot-press has been shown to result in performance increase in hydrogen fuel cells by mitigating the kinetic and ohmic losses that shape the polarization curve [3,6,9]. Power is a key figure of merit in energy conversion systems and portrays the rate at which energy is produced, where the current density and potential are simply multiplied together [19]. Fig. 1 shows our polarization testing of Nafion 212 (N212) and Nafion 115 (N115) membranes at conditions described in Experimental section 2.1 before and after 3 min of hot-press time. Gains in maximum power density can be seen in both membranes after the hot-press procedure. The maximum power increased 143% from 206 to 500 mW cm^{-2} for N212. The maximum power increased 42% from 287 to 408 mW cm^{-2} for N115. However, the power increase in the Nafion 212 is disproportionately larger due to hot-press than the increase in the Nafion 115. This could be due to the differences in the behavior of the membranes under thermal and compressive stress. For example, the tensile strength of N212 is 23.9 MPa [20] and 29 MPa for N115 [21], which implies that N212 may flow more easily and better reconfigure under hot-press (HP) [22]. Higher potential is evident in the polarization plot. Using a reference point of 400 mA cm^{-2}, the HP N212 has a potential of 0.727 V compared to 0.486 V before hot-pressing, a gain of 49.6%. At the same reference point for N115, the HP has a potential of 0.706 V compared to 0.636 V before hot-pressing, a gain of 10.9%. Additionally, operation is sustainable at a higher current in both instances.

3.2. Effects of hot-press on the Tafel response on N212 and N115 based cells

In order to extract information about performance losses in the activation polarization dominated region, a correction is made for IR loss to get the Tafel plot, which focuses on kinetics losses (Fig. 2). It is believed that the increased contact from hot-press results in a greater number of available reaction sites that improve the kinetics of a cell, [3,6,19] and this improvement is shown in our measurements. For example, at a current density of 100 mA cm^{-2}, which is taken to be within the kinetic domain, the IR-free potential of the N212 cell after hot-press increased 2.9% to 0.812 V compared to a corrected potential of 0.790 V before hot-pressing. With the same reference point, the IR-free potential of N115 with hot-press increased 2.6% to 0.847 V, compared to a corrected potential of 0.826 V before hot-press. These slight improvements in IR-free potential can be attributed to reduced losses in the activation polarization region.

Each Tafel plot can be modeled using Butler–Volmer kinetics and mass transport effects as defined by Bard and Faulkner [23].

$$V_{\mathrm{IR-Free}} = E_a - b\log(i) + C\log\left(\frac{i_{\lim} - i}{i_{\lim}}\right) \tag{3}$$

where

$$E_a = E_{\mathrm{theor}} + b\log(i_0) \tag{4}$$

These equations define b as the Tafel slope, C and $i_{\lim}$ as mass transport limiting conditions and i is the operating current. E_{theor} is the theoretical, open circuit potential and i_0 is the exchange current density and describes the reaction rate at the electrode surface [24].

The IR corrected potential becomes consistently larger once cells have become hot-pressed (Table 1). Higher Tafel slopes can be

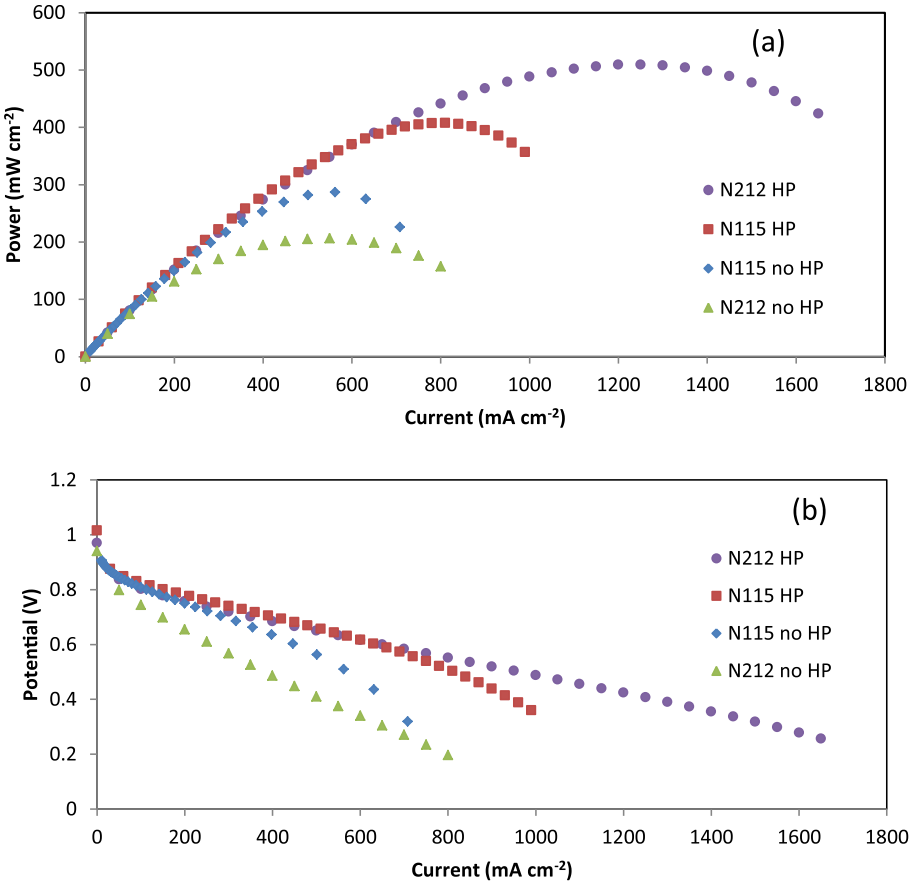

Fig. 1. Power (a) and polarization (b) curves for N115 and N212 before and after 3-min hot-press. Tests were performed under the following conditions: 60 °C cell temperature, 100 sccm flow rate for H_2 and O_2, 15 psi back pressure.

attributed to kinetic loss which implies that the hot-press procedure improves the kinetics of the cell. Fig. 2 and Table 2 display a larger benefit to hot-pressing of the Nafion 212 MEA. From these parameters, a 43.8% decrease in the Tafel slope occurs for the N212 hot-press which is greater than the 20.4% decrease in the slope of N115. The improved slope and higher value of the i_{lim} parameter, which indicates the improved mass transport of the N212 membrane, together improve the performance of the N212 HP above the N115 HP.

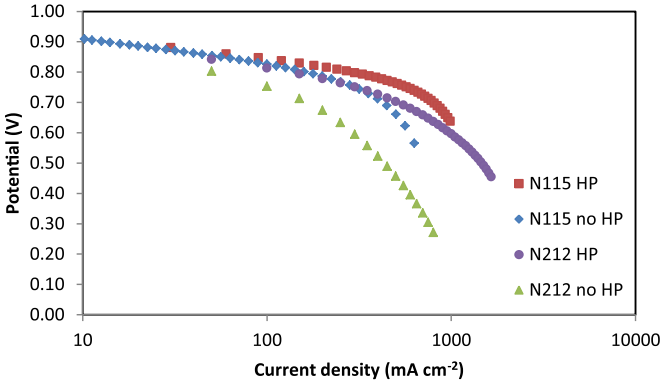

Fig. 2. Tafel Plot of IR-corrected potential vs. current density for N115 and N212 before and after 3-min hot-press. Tests were performed under the following conditions: 60 °C cell temperature, 100 sccm flow rate for H_2 and O_2, 15 psi back pressure.

Because we press at the glass transition temperature, both flow of polymer membrane material and flow of ionomer in the catalyst layer are possible. Nafion can flow outward to fill voids between the membrane and GDL. Meanwhile, ionomer can travel toward the interface to fill these voids. Additionally, electrically isolated Pt/C can be bridged during catalyst reconfiguration, allowing for electron travel from previously inactive sites. Even though reconfiguration can improve ohmic contact and catalyst utilization, excessive hot-press can hinder performance through the loss of water content, and drying and degradation of the membrane. In Fig. 2, hot-press shows results favorable to reconfiguration. Voids can arise more easily in Nafion 212 since conformal contact during assembly is slightly more difficult with the thinner membrane. Therefore, there is a greater potential for improvement when hot-press occurs in Nafion 212 since Nafion 212 has a better ability to reconfigure than Nafion 115. Positive results can be attributed to a better

Table 1
Data gathered before and after 3-min hot-press. Tests were performed under the following conditions: 60 °C cell temperature, 100 sccm flow rate for H_2 and O_2, 15 psi back pressure.

Membrane	Max power density (mW cm^{-2})	Potential (V)@400 mA cm^{-2}	IR corrected potential (V)@400 mA cm^{-2}	R_{Ohm} (ohm cm^2) @400 mA cm^{-2}
N115 no HP	287	0.636	0.826	163
N115 HP	408	0.706	0.847	195
N212 no HP	206	0.486	0.790	92
N212 HP	500	0.727	0.813	103

Table 2
Kinetic and mass-transport parameters given by Equation (3) at discussed hot-press specifications. Tests were performed under the following conditions: 60 °C cell temperature, 100 sccm flow rate for H_2 and O_2, 15 psi back pressure.

Tafel	E_a (mV)	b (mV decade^{-1})	C (mV decade^{-1})	iL (mA cm^{-2})
N115 HP	769	74	107	1030
N115 no HP	730	93	244	802
N212 HP	708	109	421	2160
N212 no HP	619	160	660	1150
N212 HPx2	714	104	1401	15,700
N212 HP 6 min	692	116	244	1770

catalytic layer configuration and improved boundary between the catalyst and membrane, with Nafion 212 showing greater improvement than Nafion 115.

3.3. Effects of hot-press on ohmic resistance behavior

Fig. 3 shows the effect of the hot-pressing on ohmic resistance. Resistance data is generated by the current interrupt method. The fuel cell test system performs this test as current is scanned. T. Ward et al. indicate that the thinner Nafion 212 membranes have lower resistance than Nafion 115, which can be seen in Fig. 3 [25]. This data suggests slightly higher resistance results from the hot-press process, likely due to the desiccation of the MEAs during the hot-pressing procedure. Therefore, other factors, including improved kinetics shown in Fig. 2, account for the improved power and potential of hot-pressed MEAs, and changes in resistance do not. The heat and pressure can also compress and lead to closure of pores within the Nafion membrane [10]. In turn, this would lead to elevated membrane resistance. However, for the testing conditions employed here, resistance increase was not significant which suggests minimal damage to the membrane as a result of hot-pressing.

3.4. Effect of repeated hot-pressing on the fuel cell performance of N212 based cells

Consecutive hot-presses with conditioning between presses for Nafion 212 increased the maximum power density over the already improved single hot-press for the same cell (Fig. 4). The maximum power density improved 160% from 209 mW cm^{-2} before hot-press to 543 mW cm^{-2} after a single hot-press. After conditioning and a second hot-press, the power density improved an additional 47% to 799 mW cm^{-2}. From zero to two presses, the potential at 400 mA cm^{-2} rose from 0.540 V to 0.676 V to 0.712 V.

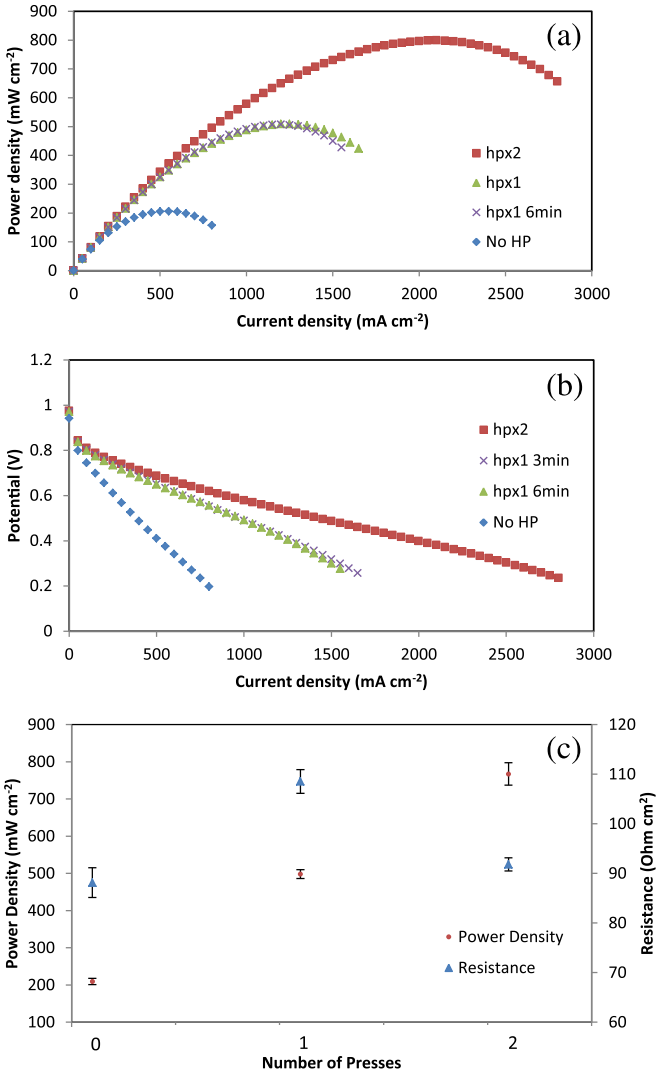

Fig. 4. Effect of repeated 3-min hot-press on Nafion 212. Tests were performed under the following conditions: 60 °C cell temperature, 100 sccm flow rate for H_2 and O_2, 15 psi back pressure.

Subsequent testing refuted the hot-press time as a contributing factor to improved kinetics. A Nafion 212 membrane was hot-pressed in a single step for 6 min to equalize the hot-press time of a double hot-pressed MEA. The results were essentially

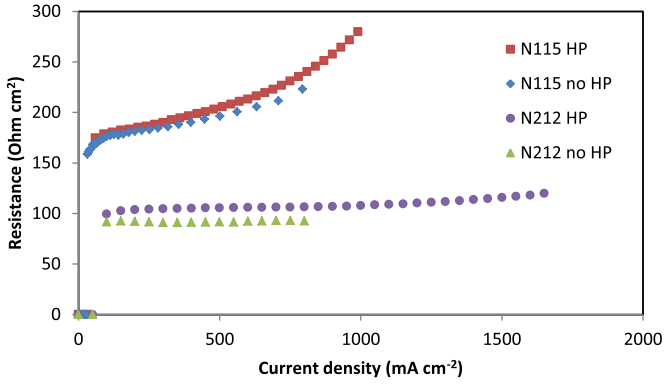

Fig. 3. Resistance effect of 3-min hot-press in Nafion 115 and 212. Tests were performed under the following conditions: 60 °C cell temperature, 100 sccm flow rate for H_2 and O_2, 15 psi back pressure.

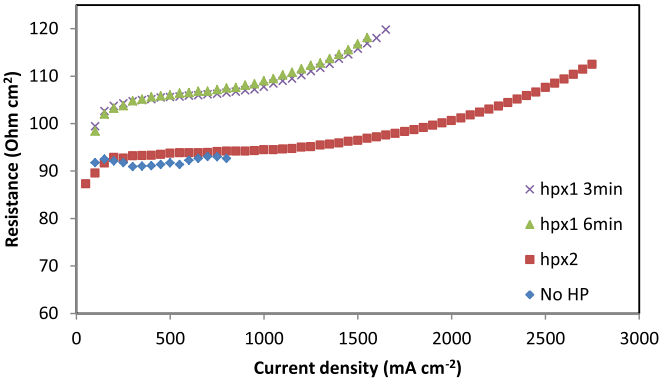

Fig. 5. Repeated hot-press effect on resistance. Tests were performed under the following conditions: 60 °C cell temperature, 100 sccm flow rate for H_2 and O_2, 15 psi back pressure.

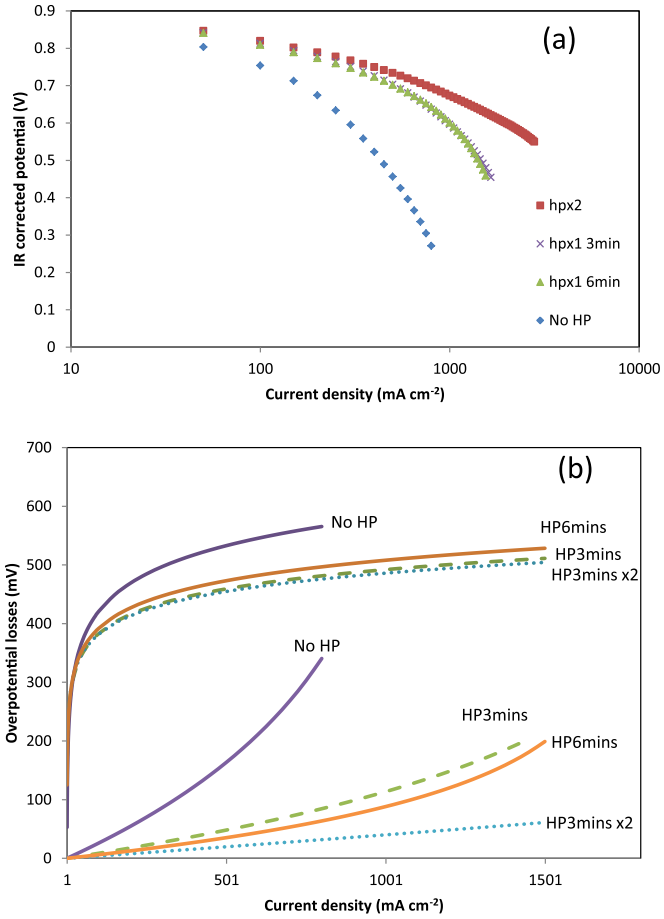

Fig. 6. Tafel plot for repeated hot-press (a) and activation and concentration polarization losses (b). Tests were performed under the following conditions: 60 °C cell temperature, 100 sccm flow rate for H_2 and O_2, 15 psi back pressure.

unchanged from the original results from a single 3-min hot-press. Max power density was within 0.2% of its original value (509 mW cm^{-2}–508 mW cm^{-2}) after the 6-min hot-press. At 400 mA cm^{-2}, the potential is almost equal at 0.676 for a 3 min press and 0.681 V at 6 min. However, the 6 min hot-press is unable to sustain higher current densities than the 3 min. This suggests that 3 min is a more optimal hot-press time for a single pressing.

3.5. Effect of repeated hot-pressing on the ohmic resistance behavior of N212 based cells

The resistance of the Nafion 212 membrane decreases from one to two three-min presses (Fig. 5). At a reference point of 100 mA cm^{-2}, the resistance decreases 14% to 87 mOhm cm^{-2} after a second press. This could be the result of increased electrical contact, which lowers interfacial resistance between MEA layers. The lowered resistance allows for lower IR loss to the potential and helps sustain use over higher current densities [19].

The additional 3 min of hot-press time within a single cycle, however, does not make a significant difference in the resistance (Fig. 5). At the reference point of 400 mA cm^{-2}, there is a 0.9% difference from 107 mOhms cm^2 at 3 min to 106 mOhms cm^2 at 6 min. Hence, the performance enhancement is not explained by increased time under hot-pressing conditions. Because conditioning was performed between hot-presses, hydration could be an important issue.

3.6. Effect of repeated hot-pressing on the Tafel response of N212 based cells

The Tafel plot is created after eliminating IR-losses (Fig. 6). At 400 mA cm^{-2}, the IR-corrected potential increases 5.6% from 0.769 V to 0.812 V for HP to HPx2. There is an improvement of Tafel slope from 109 mV $decade^{-1}$–104 mV $decade^{-1}$ for HP to HPx2 (Table 2). As Fig. 6 is corrected for IR-loss, this suggests that the benefits from the double hot-press procedure extend to the kinetic and concentration polarization domains.

In order to separate and better explain the effects of activation and concentration polarization, the parameters in Table 3 were used to construct Fig. 6b. Clearly, the hot-pressed MEAs exhibit superior kinetic and concentration overpotential suppression attributes. At a high current density of ~800 mA cm^{-2}, there is ~90 mV difference between the double hot-pressed cell and the un-pressed one. The spread in the activation overpotential data for all the pressed cells is ~24 mV (~5%). Fig. 6b also shows greater concentration polarization losses in the un-pressed MEA. The effect of hot-pressing translates through all the layers and materials that make up the MEA including the gas diffusion layer GDL, microporous layer (MPL), membrane, and catalyst layer (CL). As hot-pressing shrinks the diffusion media and leads to the formation of thinner MEA structures with shorter diffusional path [26], its benefits are seen in all the hot-press treated MEAs. However, hot-pressing can also lead to the closing of pores and increasing gas-phase transport resistance [10]. We conclude that the double press procedure resulted in a more optimized configuration, capable of mitigating transport losses.

3.7. Effect of repeated hot-pressing on the water content of N212 membranes

The water content indicates the degree to which Nafion 212 can hydrate after being treated to various conditions. Previously it has been shown and we have verified above that Nafion 212 increases performance after one 3-min hot-press [3,6] and slightly declines its power output with an additional 3 min of hot-press time (in a single cycle). In order to correlate water content, Nafion strips were subjected to the previously described conditions: no hot-press, 3-min hot-press, 6-min hot-press, and a double 3-min hot-press with 2-h water soak between presses to simulate conditioning. As

Table 3
Repeated hot-press data at discussed hot-press specifications. Tests were performed under the following conditions: 60 °C cell temperature, 100 sccm flow rate for H_2 and O_2, 15 psi back pressure.

# of HP	Repeated hot-press max power density (mW cm^{-2})	Potential (V) at 400 mA cm^{-2}	Fig. 5 Resistance (ohm cm^2)@400 mA cm^{-2}	Fig. 6 IR corrected potential (V)@100 mA cm^{-2}
0 min	219.2	0.540	88	0.790
1 × 3 min	509.3	0.676	109	0.819
1 × 6 min	507.5	0.681	106	0.809
2 × 3 min	799.4	0.712	92	0.812

indicated in Fig. 7, no hot-press produced a water content of 17.4% with a standard deviation of 0.6%. A 3-min hot-press produced a water content of 19.9% with a standard deviation of 0.4%. The water content of the 6-min was 15.3% with a standard deviation of 1.4%. The double hot-pressed membrane produced a water content of 20.9% with a deviation of 2.0%.

There is a correlation between water content and our previous results. Using ± one standard deviation as a measure of uncertainty, we can conclude that there is a higher water content for both the 3 min and double hot-press than the 0 min and 6 min hot-pressed Nafion. The membrane that was not pressed had a higher water content than the 6 min pressed membrane. This could signify that thermal damage that hinders hydration starts to take place after the optimal hot-press time of 3 min and agrees with Yildirim's findings of lowered swelling degree for cells that have had 6 min of heat treatment [27]. The 6-min hot-pressed cell had a similar performance to the 3-min hot-pressed cell, which may signify the competition between increasing electrical contact and drying out of the membrane. In agreement with our data, water content maximizes at both 3 min and for the 3-min double hot-pressed cell. Though the double hot-pressed cell has a higher average value, we cannot conclusively determine it has a higher water content within one standard deviation range. We can therefore conclude that hydration could be a contributing factor to fuel cell performance in hot-press processes and merits further investigation. Increased electrical contact and a greater ability to store water and transport hydronium ions optimize the performance of the 3-min and 3-min double hot-pressed cell. Earlier data (Fig. 3) presented a slight increase in resistance with hot-press which does not significantly affect these results.

Fig. 4 shows that repeated hot-press provides the best performance of membranes with different hot-pressing variables. All three major losses in the polarization are mitigated through the double hot-press procedure. A combination of improved Tafel slope, lowered resistance, improved concentration polarization, and increased water content, allow for the heightened performance of double hot-pressed cells. We suspect that improved Tafel slope, reflective of reduced kinetic overpotential, most greatly benefits the repeated hot-pressed cell from a single hot-pressed cell. The reduced resistance (Fig. 5) likely due to increased water content also benefits the cell immensely. Improved concentration polarization allows for higher sustained current densities and also raises performance significantly.

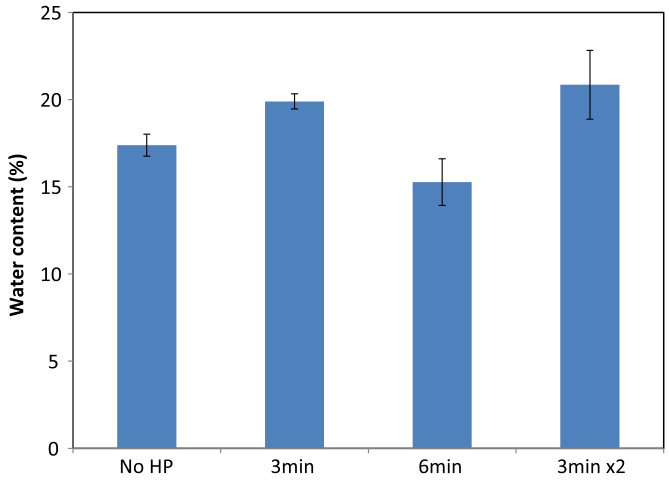

Fig. 7. Water content at discussed hot-press specifications. In Table 4, dry weights were recorded after drying for 3 days at 80 °C, and wet weights were recorded after the 2-h, 80 °C water soak and 5 day, room temperature water soak process.

Table 4
Water content at discussed hot-press specifications. Dry weights were recorded after drying for 3 days at 80 °C, and wet weights were recorded after the 2-h, 80 °C water soak and 5 day, room temperature water soak process.

Dry measurements		Wet measurements	
Sample	Average (mg)	Average (mg)	Water content (%)
s1	31.55 ± 0.08	37.04 ± 0.18	17.40
s2	34.06 ± 0.07	40.84 ± 0.12	19.91
s3	35.07 ± 0.06	40.43 ± 0.46	15.28
s4	32.98 ± 0.05	39.86 ± 0.65	20.86

4. Conclusion

The effect of hot-press and repeat hot-press on fuel cell performance was studied. Maximum power increased by 117% for Nafion 212 cells after one hot-press and showed an additional 36.4% increase in potential. Similar gains can be seen in Nafion 115 cells. These gains are not strictly attributable to IR losses as shown by the decrease in Tafel slope. With conditioning preceding a second hot-press, an additional 47% increase in maximum power density, and 5.4% increase in potential occurred. These procedures were shown to lower the resistance of the cell and increase its ability to function at high current densities.

Hot-press duration was excluded as a contributing factor for these increases, and a 3-min optimal hot-press time is confirmed. Using one standard deviation uncertainty for water content data, a correlation between our results and hydration is evident. Future testing will determine if additional hot-presses can improve performance further. Possible MEA damage from excessive hot-press cycles and hot-press time will be evaluated.

Acknowledgments

The authors are grateful for the support of the Army Research Office under Grant No. W911NF1010453.

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